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Modeling spatiotemporal variability of intra-urban air pollutants in Detroit: A pragmatic approach

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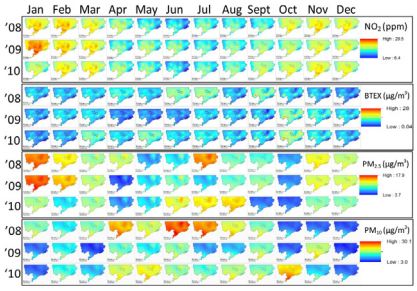
HIGHLIGHTS

- This method integrates temporally and spatially detailed air quality datasets.
- NO_2, BTEX, $PM_{2.5}$, and PM_{10} were investigated.
- A series of monthly concentration models was generated for the Detroit airshed.
- Temporal trends and neighborhood scale spatial variability were preserved.

GRAPHICAL ABSTRACT



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ABSTRACT

This study combined a three-year time series of air pollutant measurements from the Michigan Air Sampling Network (MASN) with spatially detailed datasets for two two-week periods in September 2008 and June 2009. The objective was to produce monthly pollutant concentration models for the city of Detroit, Michigan, USA from January 2008 through December 2010, in support of a related epidemiological study examining adverse birth outcomes in Detroit. Two gaseous analytes, NO_2 (nitrogen dioxide) and total BTEX (benzene, toluene, ethyl-benzene, and xylene), as well as two particulate matter size fractions, $PM_{2.5}$ and PM_{10}, were investigated. The September 2008 and June 2009 datasets were modeled using ordinary kriging to produce high spatial density concentration maps with 300 m by 300 m resolution across the city. A weighted average was applied to these maps to generate a series of monthly spatial models for each pollutant. Temporal variability was then incorporated by adjusting each monthly spatial model using an average bulk shift derived from MASN time series measurements for the corresponding month over the three-year study period.

The resulting models incorporate temporal trends while preserving neighborhood scale spatial variability. Seasonal variation was evident in NO_2 models, but not readily discernable in BTEX or PM models across the three year study period. The greatest spatial and temporal variability was observed in the BTEX distributions, which are inferred to be strongly influenced by local sources. The methodology employed assumes that the interpolated monthly models adequately capture spatial variability of the air pollutants across the study area, the spatial distribution of pollutant concentrations remained consistent while their magnitude fluctuated from month to month, and that the available time series measurements reflect temporal trends across the city of Detroit throughout the three-year study period.

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1. Introduction

Urban airsheds are heterogeneous and air pollution concentrations in urban areas vary over space and time (Kim et al., 2005; Pinto et al., 2004). As a result, epidemiological studies relating health outcomes to air pollution require both spatially and temporally resolved air pollutant models to estimate acute and chronic exposures. Distributed and prolonged air quality measurements are resource intensive, however, and study designs frequently balance tradeoffs between spatial and temporal resolution (Beevers et al., 2013). Consequently, there is a growing need to develop practical methods to integrate detailed spatial and temporal air quality data from multiple sources (Mayer, 1999; Ross et al., 2013; Wilson et al., 2005).

Air sampling networks established to monitor compliance with national ambient air quality standards (NAAQS) are important sources of outdoor air quality information in the United States. In Michigan, the Michigan Air Sampling Network (MASN) uses strategically placed monitors to assess air pollutant levels throughout the state (MDEQ, 2013). Comparable air sampling networks in other states and countries provide long-term air quality measurements that may be used to estimate exposure for surrounding communities (e.g., Dockery et al., 1993; Pope et al., 2009; Samet et al., 2000; Zanobetti et al., 2003). Although these regulatory monitoring networks provide valuable time series measurements, they commonly lack the spatial resolution needed to provide neighborhood-scale exposure estimates (Baxter et al., 2013; Ozkaynak et al., 2013; Sarnat et al., 2013; Wilson et al., 2005).

Alternatively, temporary networks of active or passive air samplers can provide a higher spatial density of measurements over limited, discontinuous periods of time (e.g., Miller et al., 2010; Ross et al., 2013). Short-term monitoring networks can be logistically difficult to implement and expensive to repeat (Cocheo et al., 2008); however, their measurements are readily incorporated into land use regression (LUR) and geostatistical interpolation (i.e., kriging) algorithms to generate pollutant concentration models at increased spatial resolution (e.g., Hoek et al., 2008; Jerrett et al., 2005a; Künzli et al., 2004; Sampson et al., 2011). LUR and kriging models share similar limitations (e.g., they require a large number of sampling sites and are not readily adaptable to changing meteorological conditions (Isakov et al., 2011)) but have different strengths. For example, LUR models can reproduce small scale features such as roadway configurations that contribute to mobile source pollutants (Mercer et al., 2011) whereas kriging smooths concentration estimates. Conversely, kriged models can provide measures of uncertainty using estimation error variance throughout the model domain (Vicedo-Cabrera et al., 2013).

The objective of this study was to create a series of spatially detailed ambient (outdoor) pollutant concentration models in support of an ongoing epidemiological investigation of associations among adverse birth outcomes and air pollutants in the city of Detroit, Michigan, USA. This study builds upon prior investigations that associated acute exacerbations of asthma in Detroit and Windsor with exposure estimates derived from spatially detailed air pollutant models covering a two-week sampling period (Lemke et al., 2013). The birth outcome investigation requires chronic exposure estimates over individual trimesters and the total duration of each pregnancy. Moreover, the births examined in the study occurred over a three year period, so that a time series of air pollutant models is needed to calculate exposures based on each mother's residential address.

This paper describes the space-time distribution modeling of nitrogen dioxide (NO_2), total benzene, toluene, ethylbenzene, and xylene (BTEX), and particulate matter with aerodynamic diameters less than 2.5 and 10 microns ($PM_{2.5}$ and PM_{10}) concentrations across Detroit during 2008–2010. Specifically, we present the methods used to combine spatially detailed models developed from measurements in an extensive temporary sampling network with temporally rich, but spatially sparse MASN measurements, along with the resulting monthly concentration models for each air pollutant during the three year period. Finally, we discuss and evaluate our assumptions about the compatibility, representativeness, and applicability of the datasets employed to spatial and temporal modeling within the Detroit airshed.

2. Data

Air pollution data for this study were derived from two sources. The first data set was developed by the Geospatial Determinants of Health Outcomes Consortium (GeoDHOC) (Miller et al., 2010; Lemke et al., 2013). The GeoDHOC conducted two two-week air sampling campaigns in Detroit, Michigan and Windsor, Ontario between September 5–20, 2008 and May 29–June 13, 2009. A total of 100 passive samplers and 50 active samplers were deployed during each sampling event (Fig. 1). Passive samplers measured NO_2, SO_2, and volatile organic compound (VOC) concentrations at an approximate spatial density of 5 km^2 per sample. BTEX compounds comprised 64% and 72% of total VOCs measured in 2008 and 2009, respectively. Active samplers measured polycyclic aromatic hydrocarbons (PAHs) and particulate matter (PM) in three size fractions at an approximate spatial density of 10 km^2 throughout both cities. Pollutant distribution models were created using ordinary kriging with a 300 m $\times$ 300 m grid spacing. Details of sampling, QA/QC, and mapping methods for the GeoDHOC data set are given by Miller et al. (2010). Analysis of the 2008 air samples demonstrated spatial variability in air pollutant distributions between and, more importantly, within Detroit and Windsor at neighborhood scales (Miller et al., 2010). Kriging variance maps, which illustrate the distribution of estimation uncertainty, are provided as supplemental information (Fig. S1) to this paper.

The second data set consisted of time series measurements at five MASN locations within the city of Detroit from 2008 to 2010 (Fig. 1) (MDEQ, 2008). Not all analytes were measured at each location (Table 1). Measurements at two nearby MASN locations outside the city (Allen Park and Dearborn) were excluded from the study because $PM_{2.5}$ and PM_{10} measurements at these sites did not differ materially in temporal trends from the Detroit station measurements during the period examined. Hence, only MASN samplers located in the city of Detroit were included in the study. Measurements at two National Air Pollution Surveillance (NAPS) monitoring stations in Windsor were also excluded because they are outside the study area.

The Detroit MASN data set includes single sampling locations for NO_2, VOCs including individual BTEX components, and PM_{10} (Table 1). The East 7 Mile location was the only active NO_2 sampling location in the study area during the study period. At this location, NO_2 is sampled continuously using automated chemiluminescence (Federal Reference Method (FRM) RFNA-0179-035) (U.S. EPA, 2013) and hourly concentrations were reported. BTEX concentrations at Southwestern High School were derived from air samples collected over a 24 h period every 12 days using SUMMA canisters. These samples were analyzed for VOCs using gas chromatography/mass spectrometry following EPA method TO-15 (U.S. EPA, 1999). PM_{10} concentrations were measured at the Southwestern High School site over a 24 h period every six days using a High-Volume Air Sampler (FRM RFPS-1287-064) (U.S. EPA, 2013).

$PM_{2.5}$ was measured at five Detroit MASN sampling locations during the 2008–2010 study period (Table 1). $PM_{2.5}$ was measured over a 24 h period using a $PM_{2.5}$ Sequential Air Sampler (Rupprecht & Patashnick Company, Incorporated Partisol®-Plus Model 2025,

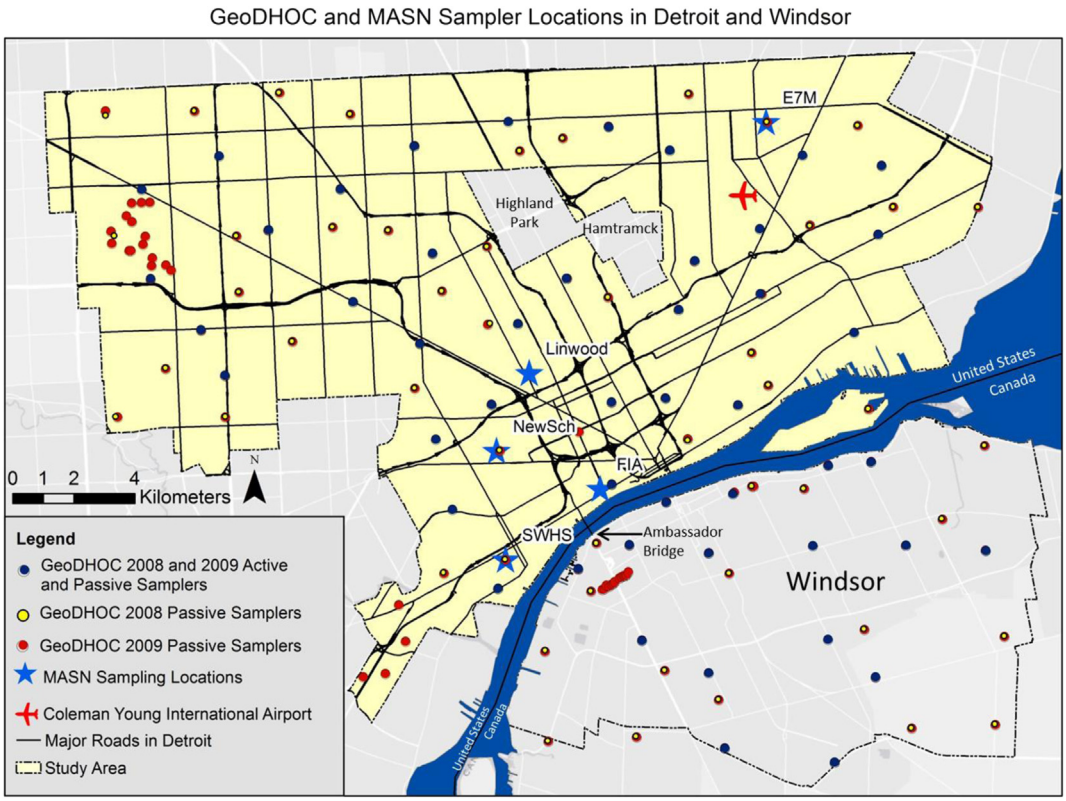

Fig. 1. Detroit study area with GeoDHOC study air sample locations in Detroit and Windsor. MASN site name abbreviations given in Table 1.

FRM RFPS-0498-118) (U.S. EPA, 2013). $PM_{2.5}$ was sampled every 3 days at each site, with the exception of the FIA/West Lafayette Street site where daily samples were available after October 1, 2009.

Additionally, hourly observations of wind speed, wind direction, temperature, and precipitation were obtained from the Coleman A. Young International Airport, located in northeast Detroit (Fig. 1) for the three-year study period. Monthly average wind rose plots (Fig. S2) indicate prevailing westerly winds throughout much of each year, although greater variability in wind direction and speed was generally present in March through May and August through September.

3. Methods

The process of integrating the MASN and GeoDHOC datasets began with a comparison of their respective measurements and model estimates. Cross validation was applied to assess the point-wise accuracy of modeled estimates. Subsequently, a two-step process was employed to develop a set of spatially and temporally interpolated concentration maps for NO_2, total BTEX, $PM_{2.5}$ and PM_{10} across the study area (City of Detroit). Initially, existing GeoDHOC maps for each pollutant were combined using a weighted average scheme to produce a series of spatially interpolated maps for twelve consecutive months. Subsequently, temporal trends derived from MASN time series measurements were superimposed on the monthly maps to generate a series of 36 monthly models spanning the period from January 2008 through December 2010 for each pollutant. Computations were performed using Surfer 11.4 (Golden Software), ArcMAP 10.0 (ESRI), and SpaceStat (BioMedware, Inc.) software.

3.1. Comparison of measurements and model estimates

GeoDHOC samplers were either collocated or placed in close proximity to three of the MASN samplers. These included the East 7 Mile location, Southwestern High School, and West Lafayette Street/FIA sites (Fig. 1). Concentrations measured by the individual GeoDHOC samplers at these three sites were compared to MASN measurements from the corresponding time periods to assess

Table 1
Michigan Air Sampling Network (MASN) monitoring sites in Detroit.

Site name	Abbreviation	Location	Analyte	Method	Sampling Frequency	Sample/Report Duration
East Seven Mile	E7Mile	Northeast Detroit	NO_2	FRM 35	Continuous	1 h
			$PM_{2.5}$	FRM 118	3 days	24 h
Linwood	Linwood	Central Detroit	$PM_{2.5}$	FRM 118	3 days	24 h
Newberry School	NewSch	South central Detroit	$PM_{2.5}$	FRM 118	3 days	24 h
FIA/Lafayette St.	FIA	South central Detroit	$PM_{2.5}$	FRM 118	1–3 days	24 h
Southwestern High School	SWHS	Southwest Detroit	$PM_{2.5}$	FRM 118	3 days	24 h
			PM_{10}	FRM 64	6 days	24 h
			VOCs	EPA TO-15	12 days	24 h

agreement between the two datasets. In addition, the spatial and temporal variability of the GeoDHOC and MASN concentration datasets were compared.

In the cross validation process, individual measured values for each pollutant were removed and re-estimated with ordinary kriging using the remaining observations. The distribution of estimation error at sampled locations was examined for magnitude, bias, and independence.

3.2. Spatial modeling

The spatially interpolated 2008 and 2009 GeoDHOC maps served as the anchor points for each pollutant (Fig. 2). These maps, which model the spatial variability of measurements integrated over continuous two-week sampling periods across the Detroit–Windsor airshed, were assumed to be representative of the spatial distribution during the month in which samples were collected (i.e., September 2008 and June 2009). A weighted average was applied to construct spatially distributed concentration models for the eight months between September 2008 and June 2009. Weighting factors for each month were assigned using fractions of the nine months separating the anchor months based on proximity in time to each anchor month. For example, the October map was blended with an 8/9 weighting for September plus 1/9 weighting for June at each point in the 300 m × 300 m model grid. Similarly, November combined 7/9 September with 2/9 June, and so on. This procedure was implemented independently for each of the four pollutants mapped.

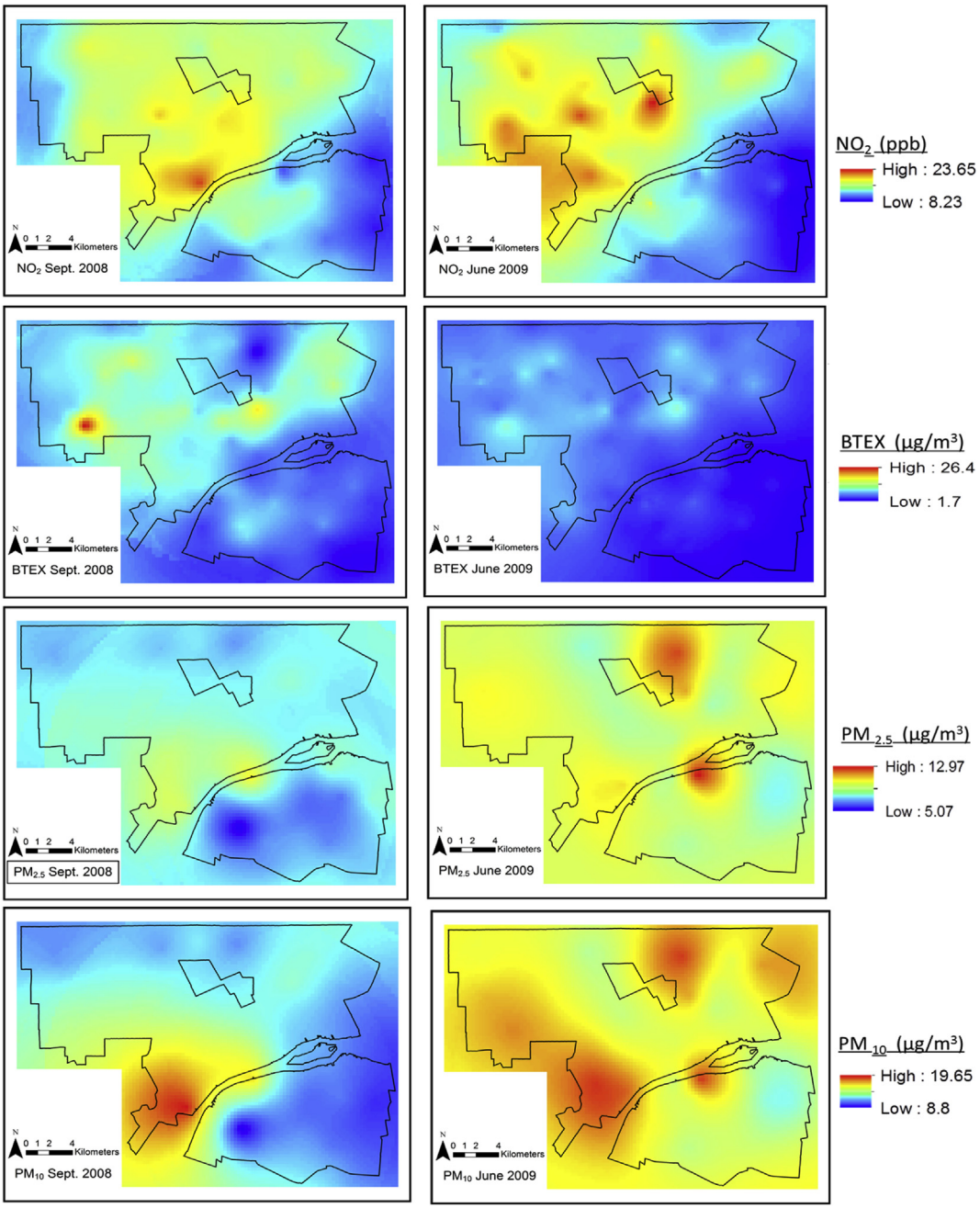

Fig. 2. Ordinary kriged maps for September 2008 (left, after Miller et al., 2010) and June 2009 (right).

July 2009 and August 2009 spatial models were also constructed using an analogous procedure to complete a 12 month series. This procedure required the additional assumption that the modeled spatial distribution of air pollutants in September 2008 can be used as a proxy for the spatial distributions in September 2009. July and August spatial distributions were calculated using a 1/3 and 2/3 weighting to combine the June 2009 and September 2009 models for each pollutant, with higher weights assigned to the temporally more proximal month. The completed year-long series of monthly GeoDHOC models (September 2008 through August 2009) served as a template of spatially variable models that subsequently were refined using available MASN time series measurements.

3.3. Temporal modeling

Temporal scaling of the monthly GeoDHOC spatial models was implemented in four steps. First, monthly averages of MASN measurements were calculated for each pollutant. This process was straightforward for NO_2, total BTEX, and PM_{10}, which were each measured at a single MASN station so that an arithmetic average of the time series measurements made during each month could be used. In contrast, $PM_{2.5}$ was measured at five MASN sites with an irregular spatial distribution throughout the city (Fig. 1). Consequently, the four $PM_{2.5}$ stations clustered in south central Detroit were first averaged together. The resulting south central Detroit value was then averaged with the East 7 Mile site in northeast Detroit to derive a crudely declustered average $PM_{2.5}$ concentration incorporating information from all five MASN sites.

Second, individual monthly GeoDHOC models (Section 3.2) for each of the four pollutants analyzed were spatially averaged over the city of Detroit, inclusive of the embedded municipalities of Hamtramck and Highland Park (Fig. 1). This process averaged the estimated values at each point in the 300 m × 300 m grid for each model.

Third, monthly spatial averages were compared to the corresponding monthly average of the MASN time series measurements. September 2008 and June 2009 are the only months in this study when direct comparisons between MASN measurements and unadjusted GeoDHOC spatial models for the City of Detroit are possible. Therefore, the mean of the September 2008 and June 2009 differences between GeoDHOC and MASN monthly averages for each pollutant was adopted as a target adjustment factor (TAF):

$$\mathrm{TAF}_i = \left[\left(\mathrm{GeoDHOC}_{\text{Sept 08}} - \mathrm{MASN}_{\text{Sept 08}}\right) + \left(\mathrm{GeoDHOC}_{\text{June 09}} - \mathrm{MASN}_{\text{June 09}}\right)\right]/2 \quad (1)$$

where subscript i represents each of the four pollutants considered.

Fourth, a spatially uniform bulk shift was calculated for each month and pollutant using the target adjustment factor and the difference between the GeoDHOC spatial model average and the corresponding MASN average:

$$\text{Bulk Shift}_{i,j} = \mathrm{TAF}_i - \left[\mathrm{GeoDHOC}_{i,j} - \mathrm{MASN}_{i,j}\right] \quad (2)$$

where subscript j represents each of the 36 months considered. When $[\mathrm{GeoDHOC}_{i,j} - \mathrm{MASN}_{i,j}]$ is positive and smaller than the target adjustment factor, positive bulk shift values are needed to increase the monthly difference up to the fixed target adjustment factor. Alternatively, if $[\mathrm{GeoDHOC}_{i,j} - \mathrm{MASN}_{i,j}]$ is positive but greater than the target adjustment factor, negative bulk shift values are needed to decrease the monthly difference down to the target adjustment factor. The converse occurs when $[\mathrm{GeoDHOC}_{i,j} - \mathrm{MASN}_{i,j}]$ is negative. The resulting bulk shift was subsequently used to adjust each monthly GeoDHOC spatial model:

$$\text{Adjusted Monthly Model}_{i,j} = \text{GeoDHOC Spatial Model}_{i,j} + \text{Bulk Shift}_{i,j} \quad (3)$$

The end product was a series of spatially and temporally variable concentration models for each of the four pollutants during each of the 36 months of 2008 through 2010.

4. Results

4.1. Comparison of measurements and model estimates

NO_2, total BTEX, $PM_{2.5}$, and PM_{10} measurements at GeoDHOC sampling locations differed from MASN values measured at collocated or nearby locations in September 2008 and June 2009. Collocated GeoDHOC and MASN measurements for NO_2 and total BTEX agreed within 25%, with the exception of June 2009 BTEX, which varied by 81% (Table 2). Although collocated PM samplers were not included in this study, GeoDHOC measurements collected within 1 km of the Southwestern High School and FIA/Lafayette St. sites agreed within 13% (Table 3).

The spatial and temporal variability of air pollutant measurements are summarized in Table S1. The magnitude of spatial and temporal variability are comparable for NO_2 and BTEX, while the observed temporal variability of $PM_{2.5}$ and PM_{10} is slightly greater than the sampled and modeled spatial variability.

For the September 2008 and June 2009 GeoDHOC models, cross validation of predicted (kriged) vs. observed concentrations demonstrates that kriging modeled estimates agree well with observed values (Table S2). Small mean errors (close to zero), small standard deviations, lack of trend in the spatial distribution of estimation errors, and the absence of conditional bias on scatterplots of error vs. estimated values indicate a lack of bias in kriged model estimations. Kriging variance maps exhibit low estimation variance throughout the majority of the study area for each pollutant (Fig. S1, Table S3).

4.2. Spatial modeling

The temporally unadjusted 12-month series of GeoDHOC models for NO_2, total BTEX, $PM_{2.5}$, and PM_{10} are presented in supplemental materials Fig. S3. Spatially averaged mean concentrations in the city of Detroit for the unadjusted September 2008 and June 2009 GeoDHOC models differed by 2–40% (Table 4). Spatially averaged mean concentrations for the 12-month series models (Table S4) vary progressively between the September 2008 and June 2009 anchor months as a consequence of the averaging method employed to construct them. Spatial variability is retained in each of the monthly models, although, in some cases, it is slightly attenuated as indicated by coefficient of variation values for individual months that are lower than the two anchor months, September 2008 and June 2009 (Table S4).

Table 2
GeoDHOC and MASN collocated sampler concentrations.

		Distance from MASN sampler	Sept. 2008	June 2009
NO_2 (ppb)	MASN (East 7 Mile)		12.1	10.3
	GeoDHOC collocated (D-P-27)	<1 m	15.5	12.5
	% Difference		24.7	19.4
BTEX (μg/m^3)	MASN (SWHS)		7.7	1.8
	GeoDHOC collocated (D-P-22)	<1 m	8.8	4.4
	% Difference		13.0	80.9

Table 3
GeoDHOC and MASN nearby sampler concentrations.

		Distance from MASN sampler	Sept. 2008	June 2009
$PM_{2.5}$ (μg/m^3)	MASN (SWHS)		11.3	9.4
	GeoDHOC (D-A-6)	990 m	10.5	10.3
	% Difference		6.9	9.1
$PM_{2.5}$ (μg/m^3)	MASN (FIA)		11.2	9.0
	GeoDHOC (D-A-30)	395 m	n/a	10.0
	% Difference		n/a	9.8
PM_{10} (μg/m^3)	MASN (SWHS)		20.4	22.0
	GeoDHOC (D-A-6)	990 m	22.5	19.4
	% Difference		9.6	12.8

4.3. Temporal modeling

Although differences were observed in $PM_{2.5}$ measurements among the five MASN locations, temporal variation tracked consistently from station to station during 2008 through 2010 (Fig. 3). Averaging the south central $PM_{2.5}$ mean value of 11.3 μg/m^3 with the East 7 Mile value of 8.5 μg/m^3 yielded a declustered Detroit average $PM_{2.5}$ value of 9.9 μg/m^3 for September 2008. Similar averaging of the south central $PM_{2.5}$ mean value of 9.1 μg/m^3 with an East 7 Mile value of 8.7 μg/m^3 yielded a value of 8.9 μg/m^3 for June 2009.

Averaged city of Detroit GeoDHOC spatial model concentrations for NO_2, total BTEX, $PM_{2.5}$, and PM_{10} also varied in relation to temporally averaged MASN measurements for the months of September 2008 and June 2009 (Table 5). Averaging of monthly differences between GeoDHOC and MASN values for September 2008 and June 2009 (Equation (1)) resulted in positive target adjustment factors of 5.2 ppb and 3.5 μg/m^3 for NO_2 and total BTEX, and negative target adjustment factors of −0.6 μg/m^3 and -6.5 μg/m^3 for $PM_{2.5}$ and PM_{10}, respectively (Table 5).

These target adjustment factors were used to calculate bulk shift values (Equation (2), Table S5) that were subsequently applied to adjust the 12-month series of GeoDHOC models for NO_2, total BTEX, $PM_{2.5}$, and PM_{10} for each month in 2008 through 2010 (Equation (3)). In some cases, this resulted in an increase in values (positive bulk shift); in others, a decrease (negative bulk shift) (Table 6). In all cases, the adjustment enforced the target difference between the MASN and GeoDHOC averages uniformly across each time series of monthly estimates for each pollutant (Fig. 4). The resulting spatially and temporally variable concentration models for each of the four pollutants during each of the 36 months incorporate the temporal trends present in the MASN data as well as the 300 by 300 m spatial resolution from the original GeoDHOC ordinary kriged maps (Fig. 5).

5. Discussion

Epidemiological studies of health outcomes related to chronic exposure to air pollutants increasingly rely upon models of urban air pollution. Practical methods are therefore needed to assimilate short-term, spatially resolved air pollution measurements with widely spaced, long-term time series data. The method presented in this paper represents a pragmatic approach used to merge two air pollutant concentration datasets with differing spatial and temporal resolution. Although we apply this method to kriged pollutant models, it could be readily adapted to scale other spatially-detailed pollutant models such as LUR. The methodology incorporates a series of assumptions and practical modeling decisions for which the justification and implications are discussed below.

To begin, our analysis incorporates air quality measurements for corresponding pollutants analyzed using different methods and varying time scales in the GeoDHOC and MASN datasets and we assume that these datasets are compatible. Similar relationships between passive sampler measurements and continuous or periodic automated fixed-site measurements have been examined in other studies. For example, Vardoulakis et al. (2009) compared chemiluminescence and passive NO_2 measurements from collocated samplers and found satisfactory agreement (relative bias and coefficient of variation < 5%) during four or five week measurement periods over thirteen months. Mukerjee et al. (2004) found BTEX measurements made with 3M organic vapor monitors over three to seven day sampling periods agreed within 10% of automated GC measurements.

In the present study, collocated GeoDHOC and MASN measurements for NO_2 and total BTEX agreed well, with the exception of June 2009 BTEX (Table 2). GeoDHOC $PM_{2.5}$ and PM_{10} concentrations measured within 1000 m of MASN stations also compared well to MASN measurements (Table 3). We emphasize, however, that GeoDHOC concentrations represent integrated measurements for two-week periods and therefore might not be expected to agree well, particularly when MASN measurements are discontinuous and infrequent during the comparative time window. For example, the June 2009 MASN monitor sampled total BTEX twice in June 2009, with 24-h reported concentrations of 2.0 μg/m^3 on June 6, and 1.7 μg/m^3 on June 18. These values agree poorly with the collocated GeoDHOC sample measurement of 4.4 μg/m^3 (Table 2). On the other hand, integrated measurements of longer duration are arguably more useful than infrequent central monitoring measurements for long term exposure estimation if the detection of peak concentrations of short duration is not essential. Given the well documented intra-urban variability of NO_2 (e.g., Jerrett et al., 2005b; Hewitt, 1991; Ross et al., 2013), BTEX (e.g., Miller et al., 2012; Vardoulakis et al., 2011), and PM (e.g., Brook et al., 1999;

Table 4
Detroit September 2008 and June 2009 GeoDHOC spatial model mean, standard deviation (SD) and coefficient of variation (CV).

		Values for Sept. 2008 Detroit	Values for June 2009 Detroit	Difference (Sept. 2008–June 2009)	% Difference
NO_2	Mean (ppb)	16.2	16.5	−0.3	2.1
	SD (ppb)	1.82	2.27	−0.45	
	CV (%)	11.3	13.7	−2.4	
BTEX	Mean (μg/m^3)	10.0	6.7	3.3	39.9
	SD (μg/m^3)	2.47	1.30	1.17	
	CV (%)	24.8	19.5	5.3	
$PM_{2.5}$	Mean (μg/m^3)	7.9	9.7	−1.8	20.2
	SD (μg/m^3)	0.53	0.72	−0.19	
	CV (%)	6.7	7.4	−0.7	
PM_{10}	Mean (μg/m^3)	13.2	16.2	−3.0	20.9
	SD (μg/m^3)	1.82	1.16	0.66	
	CV (%)	13.8	7.1	6.7	

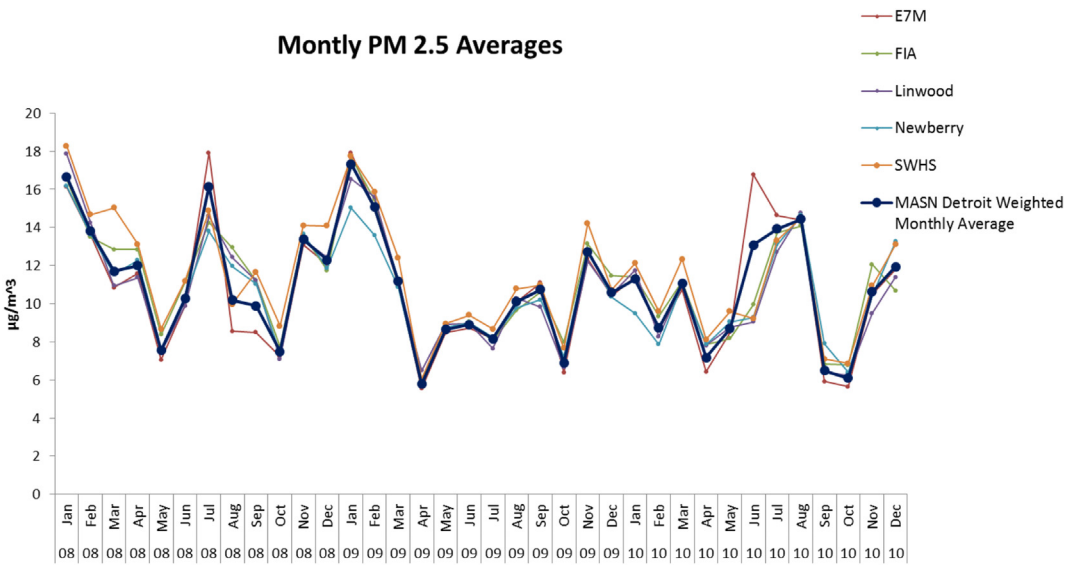

Fig. 3. MASN Monthly average $PM_{2.5}$ values at five individual monitoring sites and declustered Detroit monthly average.

Table 5
Comparison of Detroit GeoDHOC spatial model averages to MASN monthly averages.

Analyte	September 2008			June 2009			Target adjustment factor (average difference)
	GeoDHOC spatial model average	MASN average	Difference	GeoDHOC spatial model average	MASN average	Difference	
NO_2 (ppb)	16.2	12.1	4.1	16.5	10.3	6.3	5.2
Total BTEX (µg/m^3)	10.0	7.7	2.3	6.7	1.9	4.8	3.5
$PM_{2.5}$ (µg/m^3)	7.9	9.9	−2.0	9.7	8.9	0.8	−0.6
PM_{10} (µg/m^3)	13.2	20.4	−7.2	16.2	22.0	−5.8	−6.5

Rodes et al., 2010; Wilson et al., 2005) measurements, we chose not to treat the MASN values as representative of the entire city. Consequently, no attempt to incorporate a systematic bias between collocated GeoDHOC and MASN measurements was undertaken in this study. Rather, computation and subsequent modification focused on comparisons between GeoDHOC model values (spatially averaged over the entire Detroit study area) and corresponding MASN measurements for which time series measurements were available.

The rationale for this approach requires that the GeoDHOC datasets and interpolated monthly models adequately capture spatial variability of the air pollutants across the city of Detroit. Available spatial information supports this provision. Experimental semi-variograms constructed with the GeoDHOC datasets were fit with variogram models (Fig. S4) incorporating ranges as small as 6 km for BTEX to as large as 30 km for PM_{10} (Miller et al., 2010). In each case, GeoDHOC samplers, ranging in spatial density from one per 5 km^2 for NO_2 and BTEX to one per 10 km^2 for PM, were spaced at intervals well below the variogram model range. Subsequent GeoDHOC ordinary kriging models, interpolated across a 300 m × 300 m grid, honor the variogram model spatial characteristics.

The approach followed here also assumes that the GeoDHOC ordinary kriging models, which are based on two-week measurements, are representative of the distribution of the mapped pollutants for the entire months of September 2008 and June 2009. We further assume that these models can be interpolated to represent spatial distributions in the ten remaining unsampled months of the year (Section 3.2) and that these inferred spatial distributions can be extrapolated throughout the three-year study period (Section 3.3). The first assumption is supported by the constancy of MASN pollutant concentration running averages calculated for each month (Figs. S5, S6, S7, and S8). Rapid, large magnitude changes in mean concentrations are not observed and therefore not expected over the course of a few weeks for any of the pollutants considered. The second assumption implies that the location and relative magnitude of stationary and mobile sources is consistent throughout the year and that meteorological conditions were similar enough to allow spatial distributions modeled in September 2008 to serve as a proxy for September 2009. Alternatively, we considered extrapolating between the months from June 2008 to September 2008, but the meteorological conditions between June 2008 and June 2009 show distinct differences in dominant wind directions, therefore making the making the September comparison more favorable. Continuity of source distribution is supported by prior studies in Windsor, a segment of the Detroit–Windsor airshed, that established significant correlations across winter, spring, summer, and fall seasons for NO_2 and BTEX (Miller et al., 2012; Wheeler et al., 2008). Seasonal correlations in the Windsor observations suggest a reasonable degree of consistency in source

Table 6
Range of monthly bulk shift values.

	Min	Max	Range
NO_2 (ppb)	−3.8	8.3	12.1
BTEX (µg/m^3)	−3.4	5. 6	9.0
$PM_{2.5}$ (µg/m^3)	−4.1	8.0	12.1
PM_{10} (µg/m^3)	−9.7	11.5	21.2

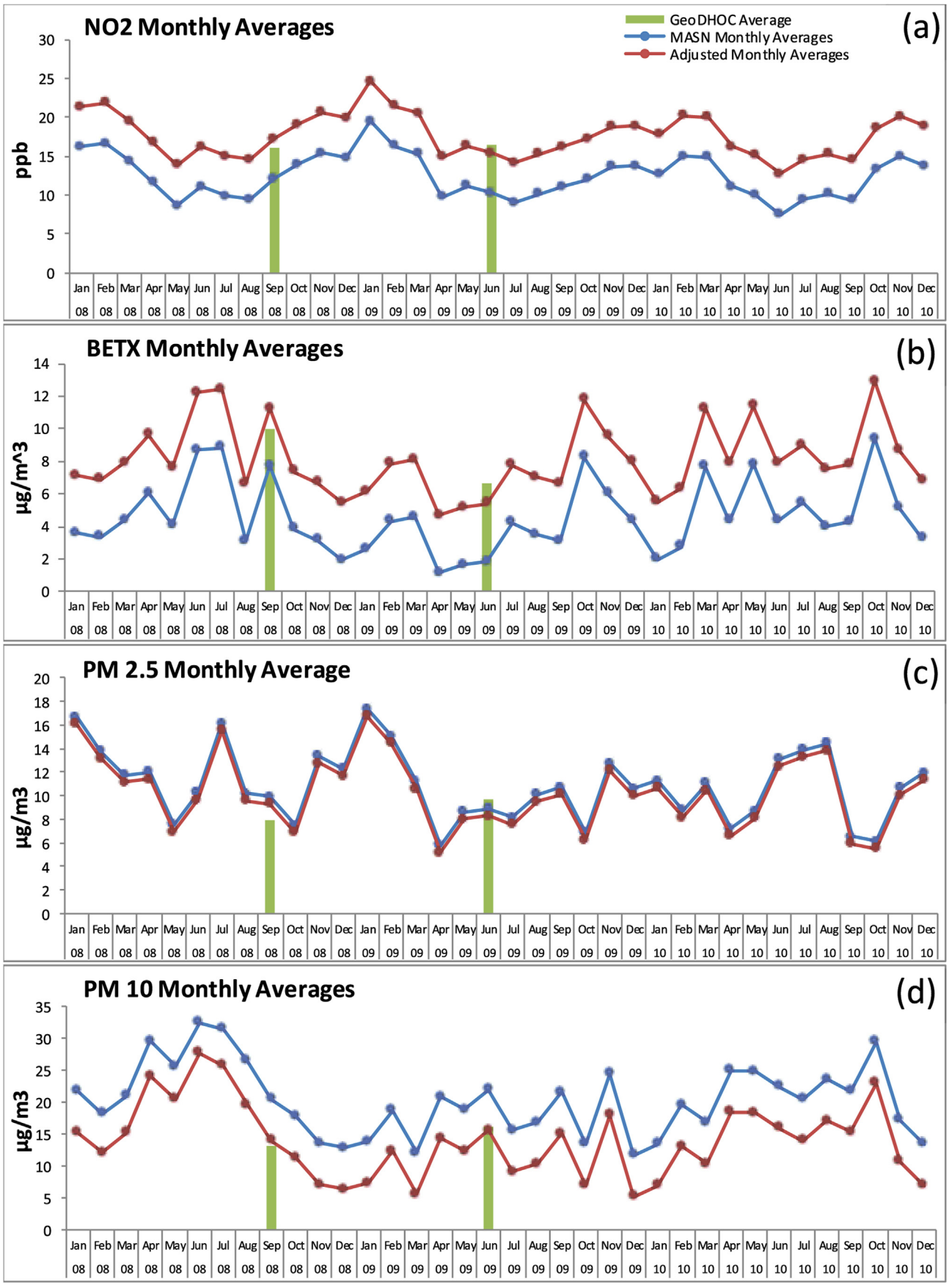

Fig. 4. MASN and adjusted monthly averages for years 2008–2010: (a) NO_2, (b) total BTEX, (c) $PM_{2.5}$, and (d) PM_{10}. Unadjusted GeoDHOC monthly averages for September 2008 and June 2009 shown as bars.

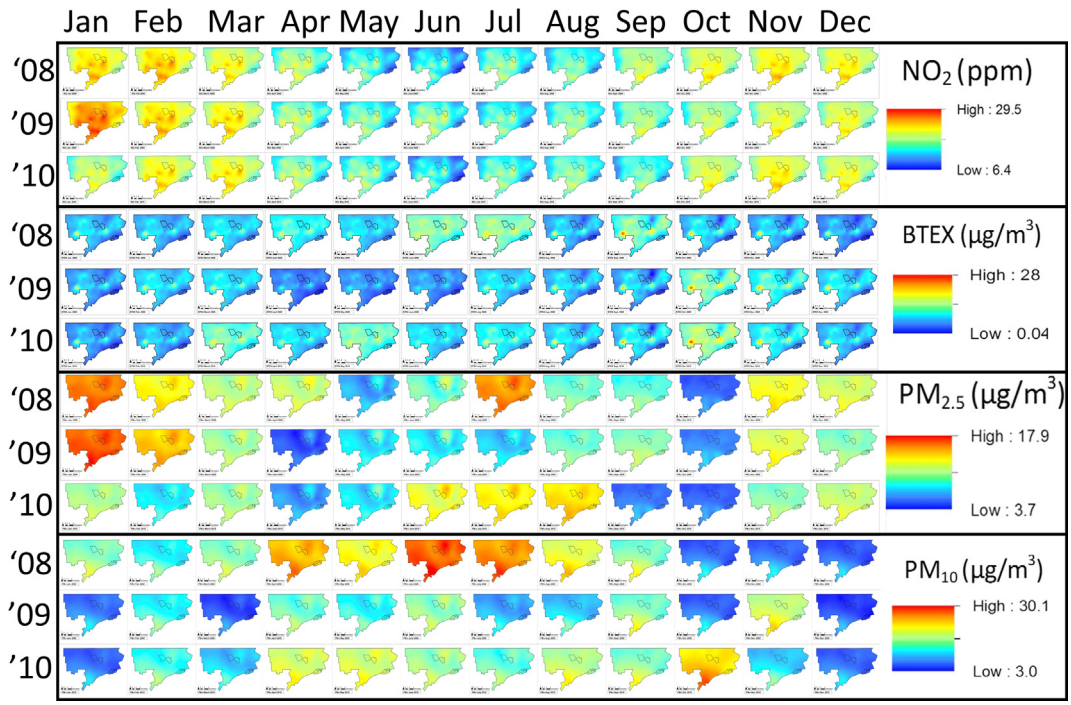

Fig. 5. Monthly NO_2, total BTEX, $PM_{2.5}$, and PM_{10} concentrations across the Detroit airshed for 2008 through 2010.

distributions, and hence spatial variability, for NO_2 and VOCs throughout the year. This supposition is more likely for pollutants derived from local sources such as NO_2, VOCs, and PM_{10}, however, than for pollutants of secondary origin like $PM_{2.5}$, which may also contain a regionally-sourced component that can depend on wind direction and other meteorological conditions.

The approach outlined here rests upon the additional assumption that the available MASN time series measurements reflect temporal trends across the city of Detroit throughout the three-year study period. Although differing measurement frequencies were employed for each analyte (Table 1), records were 95% or more complete at each MASN site during 2008–2010. In the case of $PM_{2.5}$, concentrations measured at five MASN locations across Detroit (Fig. 1) tracked consistently with each other during the three-year period of interest (Fig. 3). Sajani et al. (2004) reported similar contemporaneous temporal trends for NO_2 measured at four different stations located throughout the urban area of Bologna, Italy. This implies that although air pollutant measurements made at widely-spaced regulatory monitoring sites may fail to capture significant spatial variability in the surrounding area (Baxter et al., 2013; Ozkaynak et al., 2013), relative changes in these measurements over time are able to reflect temporal trends affecting the larger surrounding urban area.

Unquestionably, such temporal trends are influenced by daily and seasonal meteorological conditions. Weather conditions were not explicitly factored into the modeling procedures employed here and the inability of kriged maps to incorporate changes in monthly average wind direction is a limitation of our approach. However, the influence of major weather changes is assumed to be reflected in the temporal trends recorded at MASN sites. Seasonal variation in concentrations is clearly evident in the 36 monthly NO_2 concentration models (Fig. 5), with higher concentrations in winter months. Consistent seasonal variations are not evident in BTEX, $PM_{2.5}$ or PM_{10} models, however (Fig. 5). We attribute the lack of seasonality in BTEX to greater variability in local VOC source distribution (as reflected in the high CV for BTEX (Table 4)). Elsewhere, other researchers have observed a lack of seasonality in PM measurements. Brook et al. (1999), for example, found significant overlap between summer and winter seasons in 24-h $PM_{2.5}$ mass concentration distributions measured in fourteen Canadian cities and Johnson et al. (2013) found that seasonal $PM_{2.5}$ models did not predict daily concentrations better than annual models in Windsor, Ontario.

Finally, in the absence of other information, we assumed that the modeled spatial distribution of pollutant concentrations remained constant while the magnitude of the concentrations fluctuated uniformly across Detroit throughout the three year study period. As a consequence of this assumption, we employed a bulk shift to translate temporal trends from the MASN time series measurements to the monthly GeoDHOC estimates (Section 3.3). Alternative shifting techniques for incorporating the temporal trend were considered, including a ratio technique employed by Ross et al. (2013) to adjust two-week spatially interpolated air pollutant concentrations to temporal trends from continuous stationary monitors in New York City. The ratio technique was rejected, however, because it resulted in localized concentration estimates far outside (in some cases two to three times higher than) the range of measured values for several pollutant models. In contrast, the bulk shift generated concentration values within the range of observed values for each pollutant (Table 6, Table S5), except in the case of negative concentration values at a small number of grid nodes in the September 2009 and September 2010 BTEX models. BTEX values in these cells were replaced with 1/2 the method detection limit (0.1 $\mu g/m^3$) to maintain physically realistic concentrations. The underlying assumption that the magnitude of pollutant distributions fluctuate uniformly throughout Detroit becomes more tenuous when extended over longer periods of time during which major changes in infrastructure or economic conditions may occur.

6. Conclusions

The approach presented here is a pragmatic method for integrating high spatial resolution measurements from a temporary monitoring network with time series measurements from fixed regulatory monitoring stations. The resulting set of monthly concentration models preserves spatial variability captured by two detailed GeoDHOC air sampling campaigns and incorporates temporal variability present in MASN data, both of which are important for all pollutants in this study. These monthly models will be used to estimate exposure to NO_2, total BTEX, $PM_{2.5}$ and PM_{10} in an ongoing epidemiological investigation of adverse birth outcomes in Detroit.

This approach can be applied to other cities where long-term time series measurements are available to supplement spatially variable pollutant distributions modeled from temporary monitoring network datasets using kriging, LUR, or other suitable methods. Collocation of temporary monitors with fixed monitoring stations can facilitate data integration, but may not be necessary if temporal trends are consistent among multiple stationary monitoring sites.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.05.010.

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